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TRANSIENT THERMAL BEHAVIOR OF
DECOMPOSING MATERIALS

PART I GENERAL THEORY AND APPLICATION TO
CONVECTIVE HEATING

Prepared by

Thomas R. Munson
Robert J. Spindler

RESEARCH AND ADVANCED DEVELOPMENT DIVISION
AVCO CORPORATION
Wilmington, Massachusetts

Technical Report

RAD-TR-61-10

Contracts AF04(647)-258 and -305

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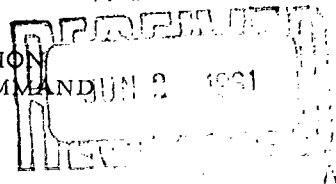
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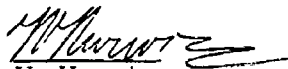
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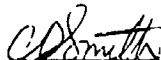
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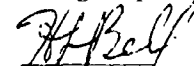
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ABSTRACT

A mathematical model is presented for the transient behavior of materials which undergo thermal decomposition. The general model accounts for a rate-controlled decomposition as well as internal heat transfer by conduction, convection, and radiation. The behavior of the model under conditions of convective heating is examined by means of analog simulation. Results of this simulation for a charring material and for Teflon are presented and the results on Teflon compared with experimental data.

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I. INTRODUCTION

Self-regulating mass transfer systems for thermal protection have received a great deal of attention in recent years primarily as a result of problems associated with the atmospheric re-entry of long-range ballistic and satellite vehicles. The status of our understanding of the ablation phenomenon has been adequately covered by Adams¹ and more recently by Hidalgo,² Steg,³ and Barriault and Yos.⁴

The major portion of the theoretical work which has appeared dealing with the ablation process has been concerned with the behavior of glassy materials.^{2, 5, 6, 7} Predictions based on these treatments have been adequately confirmed by flight test. No completely satisfactory treatment of ablation in materials which undergo thermal decomposition has appeared in the literature. The treatment of charring materials by Barriault and Yos⁴ is of limited applicability and no calculations appear to have been made using their model. Treatments of non-charring materials such as polytetrafluoroethylene (Teflon) have either considered the material response as incidental⁸ or have treated the material as a purely conductive problem.³

The purpose of this paper is to present a mathematical model of the ablation process applicable to both charring and non-charring materials. The transient behavior of both types of materials are examined briefly to indicate the utility of the model in problems of plastic ablation under conditions of convective heating.

II. MATHEMATICAL MODEL

When subjected to environmental conditions severe enough to cause material removal, some plastics (such as phenolic nylon) form a more or less adherent charred layer on the heated surface. This char is the end product of the thermal degradation process which proceeds during the heating process. Some plastics such as Teflon do not appear to form a char under any practical set of conditions. For the purposes of the present paper these non-charring materials are considered to be a limiting case of the more general behavior exhibited by the charring materials.

Conceptually the original plastic is considered as a hypothetical mixture consisting of an irreducible "char matrix" (subscript 2) and a completely decomposable "active material" (subscript 1). This active material is assumed to undergo thermal decomposition yielding only gaseous products. This decomposition is assumed to follow a first order rate law and the rate constant is assumed to vary with temperature in accordance with a simple Arrhenius expression. Since the material has continuously variable properties throughout it has not been necessary to define a "charring temperature" as in Barriault and Yos⁴ or even a char layer as such. This feature of the present model has definite advantages, both conceptual and computational, over a model requiring charring temperatures as basic input.

Consider a control volume within a bar of material whose surface is receding at an unsteady rate $\dot{s}(t)$. Figure 1 shows the geometry and coordinate system employed. Choosing unit cross-sectional area for the bar, the volume between the planes x and $(x + dx)$ is simply dx . Gaseous decomposition products are produced in unit volume at a rate $(-\dot{\rho}_1)$. This is the rate at which the active component of the material is being converted to gaseous products. It is assumed that the gases thus formed are free to move in the direction of negative x and that thermal equilibrium exists between the gaseous and condensed phases at all points in the material. Denoting by w the mass of gas flowing across unit area per unit time, continuity requires that equation (1) be satisfied.

$$\frac{\partial w}{\partial x} = \dot{\rho}_1 \quad (1)$$

The total heat, dQ , added to the control volume in time dt is the sum of the contributions due to thermal conduction, dQ_k , by gas flow, dQ_g , by chemical reaction, dQ_c , and by the absorption of radiant energy, dQ_r .

$$dQ = dQ_k + dQ_g + dQ_c + dQ_r \quad (2)$$

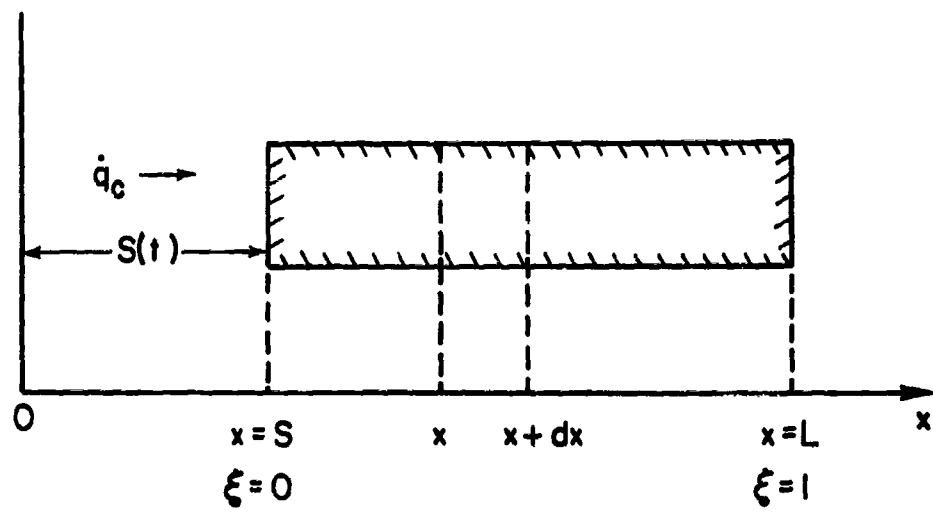


Figure 1 GEOMETRY AND COORDINATE SYSTEM

Considering the one dimensional case, these contributions are given by equations (3) through (6):

$$dQ_k = \frac{\partial}{\partial x} \left(k \frac{\partial T}{\partial x} \right) dx dt, \quad (3)$$

$$dQ_g = \frac{\partial}{\partial x} (w h_g) dx dt, \quad (4)$$

$$dQ_c = \dot{\rho}_1 \Delta H_c dx dt, \quad (5)$$

$$dQ_R = - \frac{\partial F}{\partial x} dx dt. \quad (6)$$

In these equations, k is the thermal conductivity of the bulk material, h_g is the specific enthalpy of the gaseous products, ΔH_c is the heat absorbed per unit mass of gases produced, and F is the radiant flux at any point in the material. Assuming a constant specific heat for the gaseous decomposition products and making use of the thermal equilibrium assumption, dQ_g can be written in the form of equation (7);

$$dQ_g = C_g \left(w \frac{\partial T}{\partial x} + \dot{\rho}_1 T \right) dx dt. \quad (7)$$

The heat added to the control volume is then given by equation (8),

$$dQ = \left\{ \frac{\partial}{\partial x} \left(k \frac{\partial T}{\partial x} \right) + C_g w \frac{\partial T}{\partial x} + \dot{\rho}_1 (C_g T + \Delta H_c) - \frac{\partial F}{\partial x} \right\} dx dt. \quad (8)$$

The heat equation is found by equating dQ to the change in enthalpy of the control volume and neglecting higher order differential products,

$$\frac{d}{dt} (\rho h) = \frac{\partial}{\partial x} \left(k \frac{\partial T}{\partial x} \right) + C_g w \frac{\partial T}{\partial x} + \dot{\rho}_1 (C_g T + \Delta H_c) - \frac{\partial F}{\partial x}. \quad (9)$$

In equation (9) ρ and h are the density and specific enthalpy of the control volume. Both the density and enthalpy are considered to be additive functions of the contributions due to the "inactive" and the "active" material present in the control volume. The contributions to average density and specific enthalpy due to the presence of gases have been neglected. Again, assuming constant specific heats the density and specific enthalpy may be written in the form of equations (10) and (11);

$$\rho = \rho_1 + \rho_2 \quad , \quad (10)$$

$$h = \left\{ \left(\frac{\rho_1}{\rho_1 + \rho_2} \right) C_{p1} + \left(\frac{\rho_2}{\rho_1 + \rho_2} \right) C_{p2} \right\} T \quad . \quad (11)$$

Combining equations (10) and (11) yields equation (12),

$$\rho h = \left\{ \rho C_{p1} + \rho_2 (C_{p2} - C_{p1}) \right\} T \quad , \quad (12)$$

or,

$$\rho h = A_1 \rho T + A_2 T \quad . \quad (13)$$

A similar line of reasoning with regard to the thermal conductivity results in equation (14),

$$k = B_1 \rho + B_2 \quad , \quad (14)$$

where,

$$B_1 = - \left(\frac{k_o - k_2}{\rho_o - \rho_2} \right) \quad , \quad (15)$$

$$B_2 = k_o - B_1 \rho_o \quad . \quad (16)$$

In these equations the subscript o refers to the original undecomposed material. The specific heat and thermal conductivity as defined by equations (13) and (14) are not temperature-dependent in the usual sense but are instead functions of the time-temperature history through their dependence upon the density ρ . The quantity ρ_2 is considered to be a constant and the quantity ρ_1 is assumed to vary in accordance with a first order decomposition law,

$$\dot{\rho}_1 = - \rho_1 \phi(T) \quad . \quad (17)$$

In equation (17) the function $\phi(T)$ is the first order rate constant as a function of temperature. This function may be represented by an Arrhenius expression such as equation (18),

$$\phi(T) = a \exp \left\{ - \frac{E}{T + C} \right\} \quad . \quad (18)$$

Combining equations (10), (17), and (18) equation (19) results,

$$\dot{\rho} = -(\rho - \rho_2) a \exp \left\{ -\frac{\epsilon}{T + C} \right\} \quad (19)$$

The radiant flux, F , appearing in equation (9) is considered due to external radiant heating and is assumed to obey an exponential decay law,

$$F = F_0 \exp \left\{ -\int_s^x a dx \right\} \quad (20)$$

In equation (20) F_0 is the incident radiant flux corrected for surface reflectivity and a is an attenuation coefficient which is a function of the temperature, density, and nature of the material and the wavelength of the radiant source. If the incident radiation is not monochromatic it is necessary to use suitably wavelength-averaged values for the surface reflectivity and attenuation coefficient. The effect of multiple internal reflections is neglected.

Using equation (20) the partial derivative appearing in equation (9) can be evaluated,

$$\frac{\partial F}{\partial x} = -a F_0 \exp \left\{ -\int_s^x a dx \right\} \quad (21)$$

The set of differential equations describing the system can then be written in the following form,

$$\begin{aligned} \left\{ A_1 \rho + A_2 \right\} \frac{dT}{dt} = & \left\{ B_1 \rho + B_2 \right\} \frac{\partial^2 T}{\partial x^2} + \left\{ C_g w + B_1 \frac{\partial \rho}{\partial x} \right\} \frac{\partial T}{\partial x} \\ & + \left\{ (C_g - A_1) T + \Delta H_c \right\} \dot{\rho} + a F_0 \exp \left\{ -\int_s^x a dx \right\} \quad (22a) \end{aligned}$$

$$\frac{\partial w}{\partial x} = \dot{\rho} \quad (22b)$$

$$\dot{\rho} = -(\rho - \rho_2) a \exp \left\{ -\frac{\epsilon}{T + C} \right\} \quad (22c)$$

Equations (22) together with the appropriate initial and boundary conditions then serve to determine the transient thermal behavior of an internally decomposing material.

III. APPLICATION TO CONVECTIVE HEATING

In application of the model to purely convective heating the last term in equation (22a) vanishes since $F_0 = 0$. The initial and boundary conditions employed in this analysis are as follows:

Initial conditions:

$$T(x, 0) = \text{constant} = T_0 ; \quad (23)$$

$$\rho(x, 0) = \text{constant} = \rho_0 . \quad (24)$$

Boundary conditions:

at $x = s$ (heated surface),

$$\dot{q}_c \left(\frac{\Delta h}{h_\infty} \right) - \epsilon_R \sigma T_w^4 = - \left(k \frac{\partial T}{\partial x} \right)_{x=s} + \rho_w \dot{s} \{ h_v + \eta_2 (\Delta h) \} + w_{x=s} \eta_1 (\Delta h) ; \quad (25)$$

at $x = L$ (rear boundary),

$$(w)_{x=L} = 0 , \quad (26)$$

$$\left(\frac{\partial T}{\partial x} \right)_{x=L} = 0 . \quad (27)$$

Equation (25) is a boundary condition which has been discussed a number of times in the literature,^{2, 3, 4} and is applicable at the stagnation point of a hypersonic vehicle. In this equation \dot{q}_c is the heat-transfer rate calculated to a cold isothermal wall, Δh is the difference between the enthalpy of air at stagnation conditions and its enthalpy at wall conditions, ϵ_R is the emissivity associated with the material, h_v is a specific enthalpy change associated with surface recession. η_1 and η_2 are transpiration or blowing parameters associated with the vapor species produced by internal decomposition and surface recession, respectively. These parameters can be obtained from a knowledge of the molecular weight of the vapor species and the boundary-layer flow conditions.²

In considering ablation process in a finite bar of material it is convenient to introduce a transformation to a coordinate system based on the receding surface. This transformation, first employed by Landau, is defined by equation (28),

$$\xi = \left(\frac{x-s}{L-s} \right) \quad (28)$$

application of this transformation to equations (22) and to the initial and boundary conditions yields the following set of equations:

$$\begin{aligned} \frac{\partial T}{\partial t} = & \left(\frac{1}{L-s} \right)^2 \left\{ \frac{B_1 \rho + B_2}{A_1 \rho + A_2} \right\} \frac{\partial^2 T}{\partial \xi^2} - \left\{ \frac{(C_g - A_1)T + \Delta H_c}{A_1 \rho + A_2} \right\} (\rho - \rho_2) a e^{-\frac{\epsilon}{T+C}} \\ & + \left(\frac{1}{L-s} \right)^2 \left\{ \frac{C_g w(L-s) + B_1 (\partial \rho / \partial \xi) - s(L-s)(\xi-1)(B_1 \rho + B_2)}{A_1 \rho + A_2} \right\} \frac{\partial T}{\partial \xi} \quad ; \quad (29) \end{aligned}$$

$$\frac{\partial w}{\partial \xi} = -(L-s)(\rho - \rho_2) a e^{-\frac{\epsilon}{T+C}} \quad ; \quad (30)$$

$$\frac{\partial \rho}{\partial t} + s \left(\frac{\xi-1}{L-s} \right) \frac{\partial \rho}{\partial \xi} = -(\rho - \rho_2) a e^{-\frac{\epsilon}{T+C}} \quad . \quad (31)$$

Boundary conditions:

$$\text{at } \xi = 0 \quad ,$$

$$\begin{aligned} \dot{q}_c \left(\frac{\Delta h}{h_\infty} \right) - \epsilon_R \sigma T_w^4 = & - \left\{ \left(\frac{B_1 \rho_w + B_2}{L-s} \right) \frac{\partial T}{\partial \xi} \right\}_{\xi=0} \\ & + \rho_w s \left\{ h_v + \eta_2 (\Delta h) \right\} + w_{\xi=0} \eta_1 (\Delta h) \quad ; \quad (32) \end{aligned}$$

$$\text{at } \xi = 1 ,$$

$$w_{\xi=1} = 0 , \quad (33)$$

$$\left(\frac{\partial T}{\partial \xi} \right)_{\xi=1} = 0 . \quad (34)$$

Initial conditions:

$$T(\xi, 0) = T_0 ; \quad (35)$$

$$\rho(\xi, 0) = \rho_0 . \quad (36)$$

In order to completely determine the behavior of the system an additional relation is required between the surface recession rate, \dot{s} , and one of the other parameters such as the temperature of the receding surface. The form of this additional relationship depends on the mechanism of material removal at the surface. This mechanism may be due to such effects as vaporization, combustion, or shear forces. Relationships applicable to melting materials have been considered by Fledderman and Hurwicz,⁷ and by Zlotnick and Nordquist.⁸ For purposes of checking out the analog computations, simple equations were written relating the recession rate to the wall temperature,

$$\dot{s} = \beta T_w^n \exp \left\{ - \frac{\gamma}{T_w} \right\} . \quad (37)$$

IV. COMPUTER SIMULATION

Equations (29) through (37) have been programmed for analog simulation of the thermal behavior of plastic materials. As examples of the two basic types of materials for which the model was developed, calculations were made on Teflon and on a hypothetical charring material. The calculations employing the charring material are of interest only in illustrating the behavior of the model.

A. CHARRING MATERIAL

The properties employed in the simulation are listed in Table 1. These properties were used in connection with a simulated satellite re-entry. Figure 2 indicates the stagnation enthalpy and cold wall heating employed for this run. Figure 3 indicates the two mass transfer rates as a function of time. Temperature profiles measured from the original surface are plotted in figure 4. The constants selected for this run allowed a "char" buildup of about 0.150 inch during the course of the trajectory. Several other simulations were made using constant heat inputs. The principal result of this series of runs was a demonstration of the extreme transient nature of the ablation process in decomposing materials.

B. TEFLON

A somewhat more detailed analysis of Teflon ablation was undertaken with the aim of verification of the model by comparison with the rather extensive experimental data.^{10, 11} Table 1 lists the Teflon properties selected for use in the calculations. The transpiration parameter η was obtained by assuming the decomposition product to be C_2F_4 and employing an experimental correlation based on molecular weight for mass injection into a laminar air boundary layer.¹⁰ The relation employed is given by equation (38) where μ is the molecular weight of the transpiring gas,

$$\eta = 0.60 \left(\frac{29}{\mu} \right)^{0.26} . \quad (38)$$

The constants for use in equation (37) were obtained by an analysis analogous to that of Steg.³

The simulations were made to correspond as closely as possible to the conditions in the experimental work of John and Recesso.¹¹ Figure 5 shows the amount ablated as a function of time for several of these runs. Figure 6 shows the calculated temperature distribution for one case as compared to the steady-state solution assuming constant properties and no internal decomposition. Surface temperatures and surface temperature gradients computed during the analog simulation agree with the experimental data of Hanst.¹²

TABLE I
MATERIAL PROPERTIES

	Charring Material	Teflon
ρ_0 (lb/ft ³)	64.4	137.3
ρ_2 (lb/ft ³)	32.2	0
C_{p1} (Btu/lb-°F)	0.34	0.30
C_{p2} (Btu/lb-°F)	0.34	0
k_0 (Btu/hr-ft-°F)	0.10	0.14
k_2 (Btu/hr-ft-°F)	1.00	0
C_g (Btu/lb-°F)	0.20	0.20
α (sec ⁻¹)	5.94×10^{12}	4.70×10^{18}
ϵ (°R)	50,000	72,900
ΔH_c (Btu/lb)	500	680
ϵ_R	0.7	0
h_v (Btu/lb)	200	680
η_1	0.5	0.43
η_2	0.5	0.43
β (in/sec-(°R) ⁿ)	1.96×10^8	12.9×10^4
n	0	0.5
γ (°R)	50,000	36,450

In order to compare the analog runs with the results of experimental ablation measurements, the results of the simulations were reduced in the same way as the experimental data. The results are presented in figure 7. The quantity plotted is the ratio of the cold wall heat-transfer rate to the rate of mass loss by the sample. The steady-state values predicted by the model at two different heat-transfer rates are plotted for comparison. As can be seen from figure 7, the transient nature of the process as predicted by the model shows excellent agreement with experiment when the experimental data are interpreted as transient rather than representing steady-state behavior.

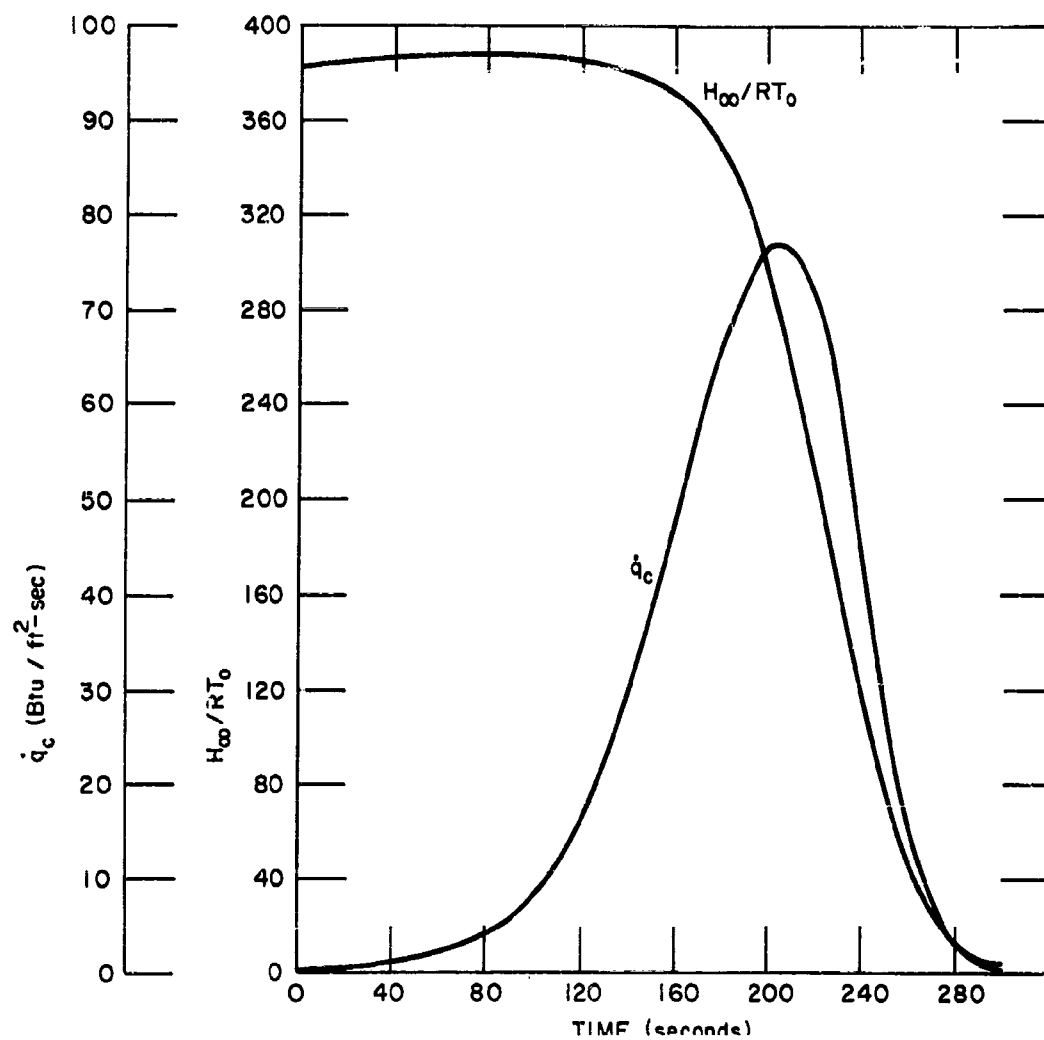


Figure 2 SATELLITE RE-ENTRY CONDITIONS

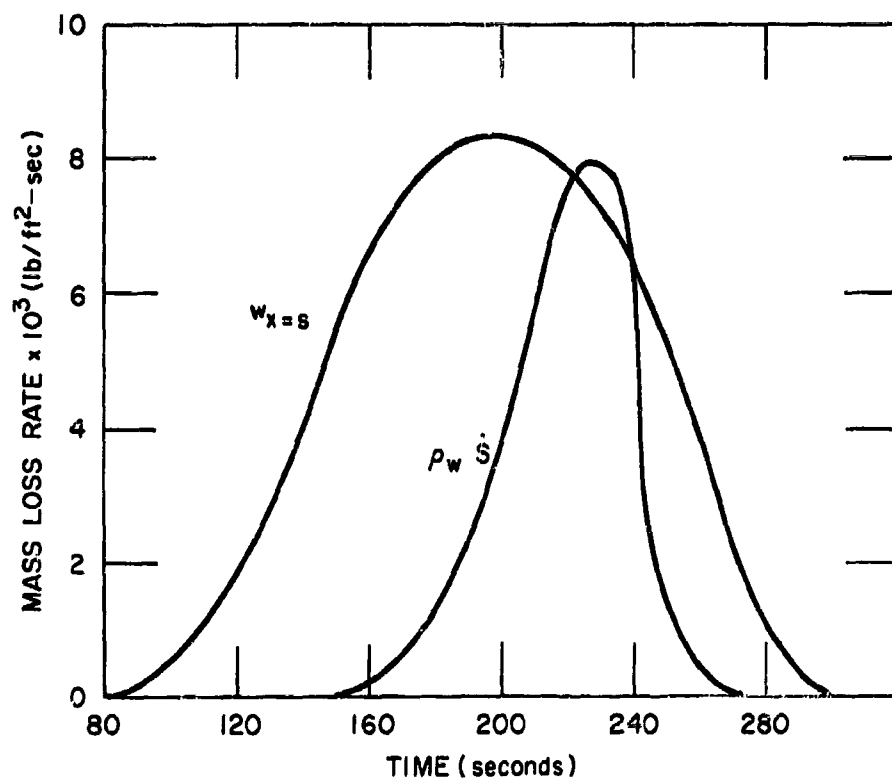


Figure 3 MASS TRANSFER RATES FOR SATELLITE RE-ENTRY
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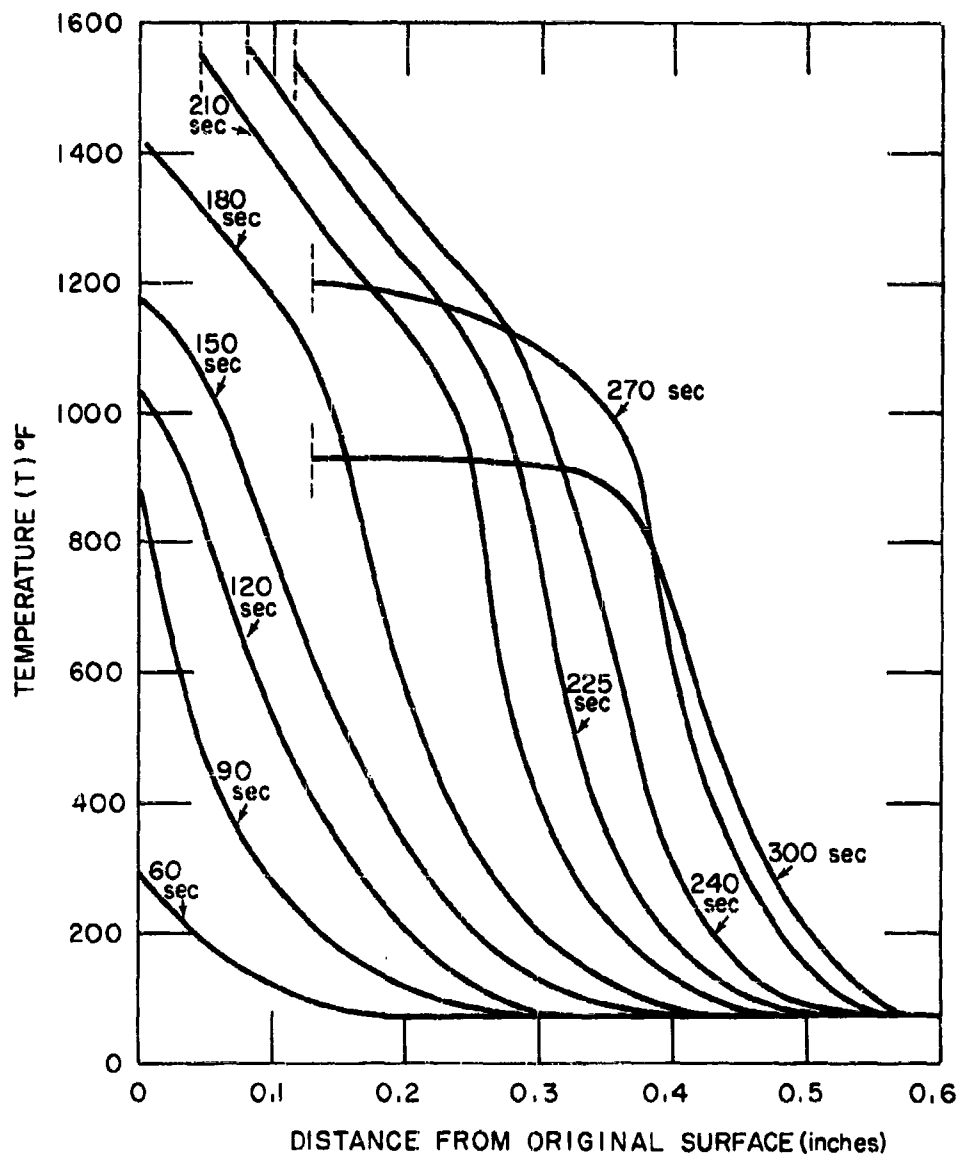


Figure 4 TEMPERATURE PROFILES FOR SATELLITE RE-ENTRY
61-1130

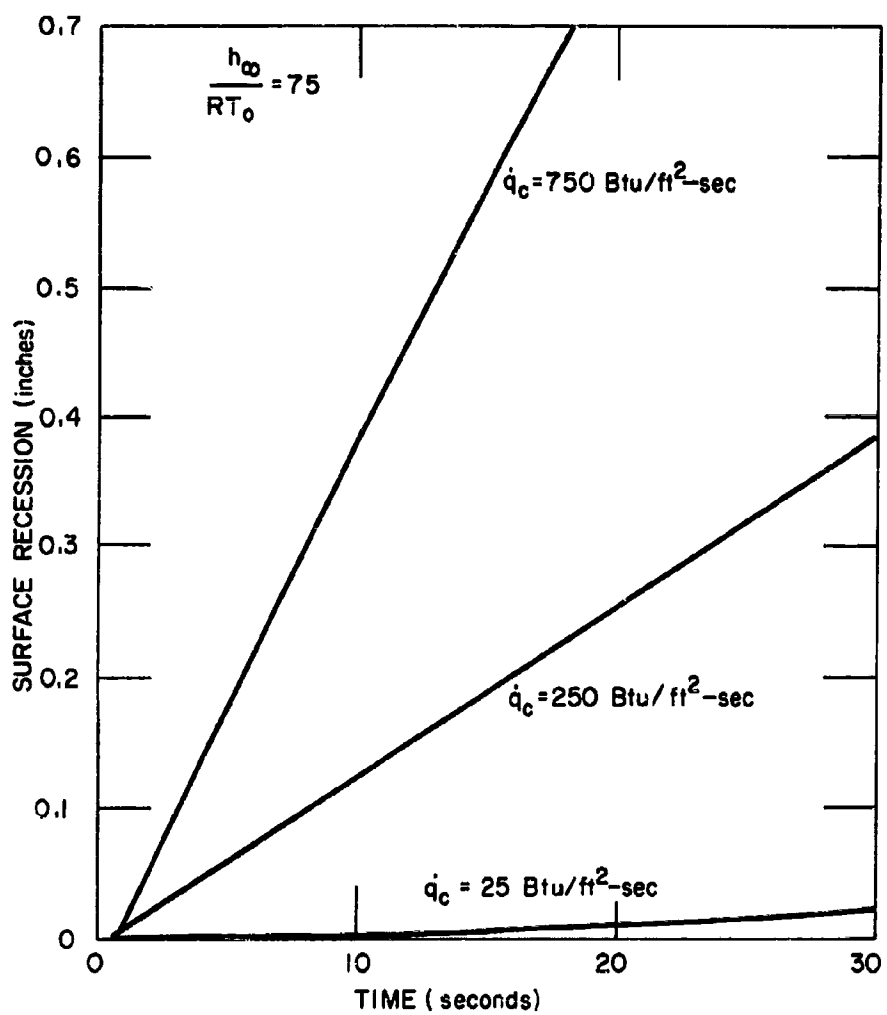


Figure 5 SURFACE RECESSION IN TEFLON
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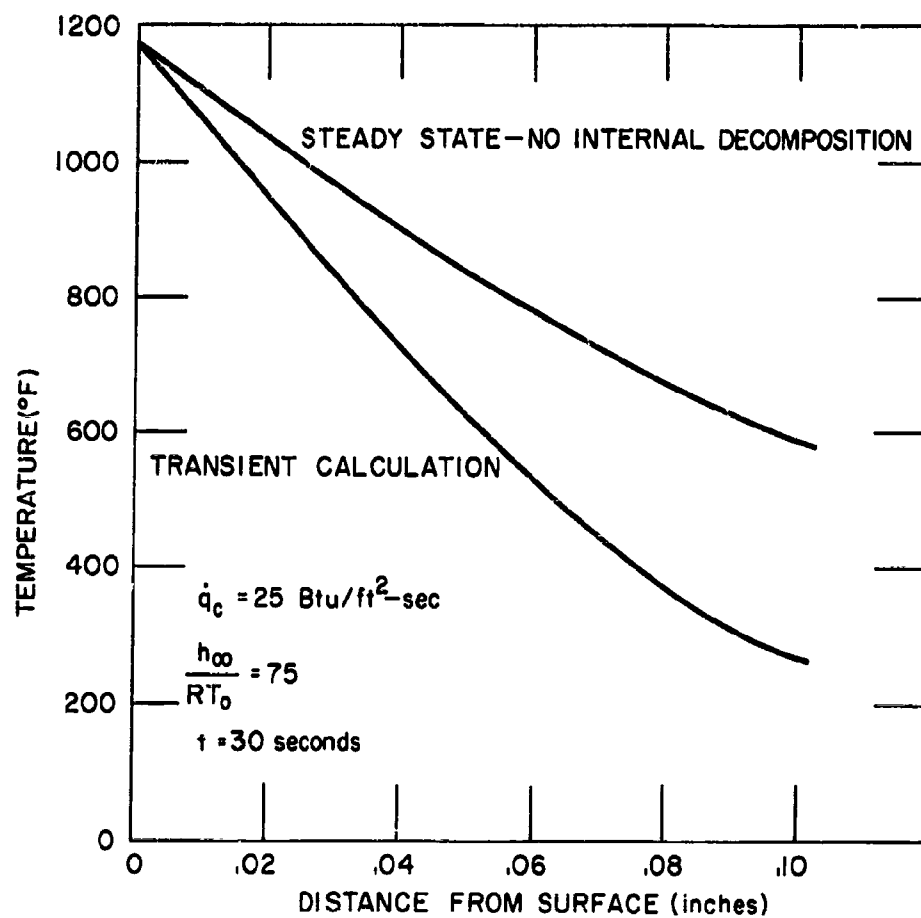


Figure 6 TEMPERATURE PROFILES IN TEFLON
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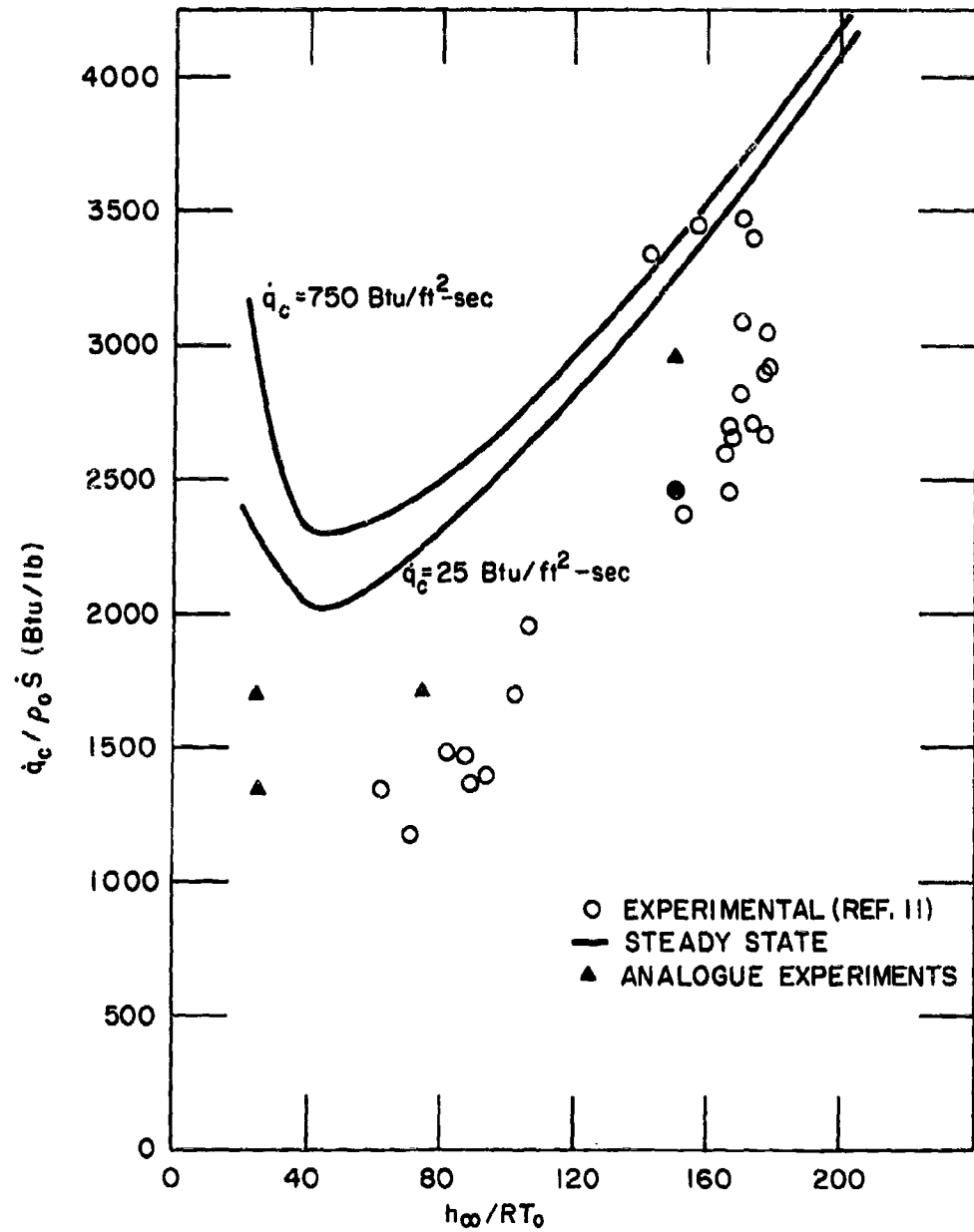


Figure 7 TEFLON HEAT OF ABLATION
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V. CONCLUSIONS

The mathematical model presented for the transient thermal behavior of materials which undergo thermal decomposition is consistent with the experimental data on these types of materials. Preliminary experience with the model indicates the necessity for considering ablation in plastic materials as a completely transient phenomenon and for interpretation of the experimental results in terms of this transient behavior.

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